Radiocarbon Reservoir Age of High Latitude North Atlantic Surface Water During the Last Deglacial

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Abstract

The radiocarbon reservoir age of high latitude North Atlantic Ocean surface water is essential for linking the continental and marine climate records, and is expected to vary according to changes in North Atlantic Deep Water (NADW) production. Measurements from this region also provide important input and/or tests of oceanic radiocarbon using 3-D global ocean circulation models. Here, we present a surface water radiocarbon reservoir age record of the high latitude western North Atlantic for the deglacial period via the use of fossil cold-water corals growing in waters that are rapidly exchanged with nearby surface waters. The reservoir age of high latitude North Atlantic surface waters was computed from the radiocarbon age difference between our radiocarbon calibration record (http://radiocarbon.LDEO.columbia.edu) and our marine radiocarbon data. 

\(^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}\) dates provide the absolute coral ages. Our high latitude North Atlantic Ocean reservoir age data combined with recalculated reservoir ages based on published coexisting terrestrial and marine material or Vedde ash radiocarbon dates from central and eastern North Atlantic shows modern values (380 ± 140 year, n = 14) during the Bolling and Allerod warm period and a 200 year increase in reservoir age (590 ± 130 year, n = 10) during the entire Younger Dryas (YD) cold episode. The reservoir age then decreased to 270 ± 20 year (n = 2) at the Preboreal/YD transition, although the dates are too sparse to be confident in this estimate. We are not able to resolve the timing of the transition to increased reservoir ages from the mid-Allerod to the YD due to the relatively small change and correspondingly large uncertainty in the estimates. The atmospheric \(\Delta^{14}C\) record derived from our atmospheric radiocarbon record displays a 40 per mil increase from 12900 to 12650 cal year BP, coincident with the shift to high reservoir ages
in the early YD cold event. Intrusion of $^{14}$C depleted Antarctic Intermediate Water (AAIW) to the high latitude North Atlantic and reduction of North Atlantic Deep Water (NADW) formation are possible causes for the coincident shift to high reservoir ages in the North Atlantic surface ocean and increased atmospheric $\Delta^{14}$C during the beginning of the YD event.

**Keywords:** radiocarbon reservoir age, high latitude North Atlantic Ocean, cold-water coral, radiocarbon calibration, Antarctic Intermediate Water (AAIW), North Atlantic Deep Water (NADW)
1. Introduction

The radiocarbon reservoir age of surface ocean water (the difference between the $^{14}$C age of the ocean surface and that of the atmosphere) reflects the balance among $^{14}$C production, the spatial variability and magnitude of $^{14}$CO$_2$ flux across the air-sea interface, oceanic circulation, and mixing with $^{14}$C-depleted intermediate and deep waters (Stuiver et al., 1991). The distribution of pre-anthropogenic (pre-1850) ocean reservoir ages grossly correlates with the main features of global ocean circulation (See http://radiocarbon.LDEO.columbia.edu for global maps of surface ocean reservoir age estimates). For the pre-anthropogenic ocean the sea surface $^{14}$C reservoir age ranges between 200 to 300 years in the subtropical gyres and increases to 1200 years at higher latitudes in the Southern Ocean and the North Pacific (Bard, 1988, Broecker et al., 1985, Butzin et al., 2005). Southern Ocean and North Pacific Ocean surface waters are “old” because of mixing with subsurface water, whose $^{14}$C concentration is lower than atmospheric values due to its extended isolation from the atmosphere and radiocarbon decay. By contrast, there is little surface reservoir age gradient in the North Atlantic Ocean between 40°N and 70°N. The radiocarbon reservoir ages of these surface waters are almost constant, at about 400 years (Bard, 1988, Broecker et al., 1985, Butzin et al., 2005) due to the North Atlantic Drift (NAD) transportation of partially equilibrated tropical and subtropical waters to high latitudes. The cooling of relatively saline surface water in the Labrador Sea and Greenland-Iceland-Norwegian Sea during winter induces active and deep (~ 2000 m) convection and contributes to deep-water formation (Lazier et al., 2002). Since radiocarbon is only produced in the upper atmosphere and transported into the surface ocean by air-sea exchange, this winter convection and deep-water
formation process rapidly mixes the surface ocean $^{14}$C content into the North Atlantic Deep Water (NADW). Broecker and Peng (1982) estimated that $\sim 164$ mol/yr atmospheric $^{14}$C is removed from the atmosphere via sinking NADW, compared to $\sim 540$ mol/yr production in the atmosphere (Masarik and Beer, 1999). Hence, records of reservoir age variation in the high latitude North Atlantic contain information about deep-water formation and ocean circulation in the high latitude North Atlantic area. Furthermore, reservoir age estimations are needed for correcting radiocarbon ages to compare marine records with continental records directly. There are several North Atlantic reservoir age estimations for the last deglacial period made possible by dating contemporaneous marine and terrestrial materials (Austin et al., 1995, Bard et al., 1994, Bjorck et al., 1998, Bondevik et al., 1999, Bondevik et al., 2006, Bondevik et al., 2001, Siani et al., 2001). A much more indirect approach is made by aligning various ice-core chronologies to $^{14}$C dated marine records by correlating the ice-core $\delta^{18}$O record and sea-surface-temperature proxy records obtained from deep-sea cores (Waelbroeck et al., 2001). However, the Waelbroeck et al (2001) study shows unusually large and rapid reservoir age variations during the last deglacial with errors that are difficult to assign due to uncertainties in ice core to marine core wiggle-matching combined with ice core age uncertainties greater than 1%. In the following discussion, we present a record of paleo-reservoir ages of the high latitude western North Atlantic Ocean by comparing the $^{14}$C ages of cold-water corals *D. dianthus* with a high resolution atmospheric radiocarbon record (http://radiocarbon.LDEO.columbia.edu). Furthermore, we combined these results with recalculated reservoir ages from contemporaneous marine and terrestrial materials from the central and eastern high latitude North Atlantic. Our results indicate close to
modern reservoir values during the Bolling and early to middle Allerod warm period. In comparison, the reservoir age was as high as 600 year during the entire YD cold interval. The reservoir age shift may result from the Northern source and southern source water mixture ratio change in the intermediate and deep water depth of North Atlantic Ocean. A benthic δ¹³C record from the high latitude North Atlantic Ocean (Rickaby and Elderfield, 2005) and atmospheric Δ¹⁴C record provide support for this interpretation.

2. Determination of high latitude North Atlantic surface water radiocarbon reservoir age

2.1 Atmospheric radiocarbon record

Our atmospheric radiocarbon record is constructed from paired ²³⁰Th/²³⁴U/²³⁸U and ¹⁴C dates on pristine surface-coral samples from offshore coral reef cores collected from Barbados (13.10°N; 59.32°W) in the western tropical Atlantic and Kiritimati Atoll (1.99°N, 157.78°W) in the central equatorial Pacific, plus tree ring (Friedrich et al., 2004) and an anchored floating tree ring record (Kromer et al., 2004) (Figure 1). The radiocarbon calibration curve is computed by a statistical model to derive a rigorous error estimation (Fairbanks et al., 2005). The 1382-ring floating tree ring record (Kromer et al., 2004) ranging from ~ 10 683 to ~12 029 ¹⁴C year BP was anchored to our surface coral record at the rapid drop of ¹⁴C ages at the beginning of YD. We applied 325 year and 335 year radiocarbon reservoir age correction to the raw radiocarbon age of Barbados and Kiritimati surface-coral samples, respectively, based on the average difference between Holocene corals and tree ring data. Annually counted tree rings range from present to 12 400 cal year BP (Friedrich et al., 2004) and corals from Barbados and Kiritimati contribute to the record from 7000 to 15 000 cal years BP, and overlap with the tree ring.
We only used the re-anchored floating tree ring record, when surface corals overlapped with the floating tree ring data set, since the floating tree ring is a more continuous chronology and has higher resolution. The consistency between the surface-coral data and tree-ring record is a direct measure of the accuracy of the coral data, and supports the assumption of near constant reservoir age for these tropical sites during the Holocene and late-deglacial period.

2.2 Marine radiocarbon record from fossil cold-water coral

Cold-water corals were recovered from Orphan Knoll (50.43°N, 46.37°W, 1600m water depth; Figure 2), located in Western North Atlantic Ocean. Today, at Orphan Knoll, the sub-surface layer between 500 and 2100 m is comprised of cold, fresh, and well-ventilated Labrador Sea Water (LSW) (Smethie and Fine, 2001). Below this parcel of LSW are Iceland-Scotland Overflow Water (ISOW) between 2100-3200 m, and Denmark Strait Overflow Water (DSOW) below 3200 m depth (Smethie and Fine, 2001). Orphan Knoll is an ideal location for estimating the reservoir age of high latitude North Atlantic Ocean surface water during the last deglacial period via paired $\text{^{230}Th}/\text{^{234}U}/\text{^{238}U}$ and $\text{^{14}C}$ measurements of the cold-water coral species $D.\ dianthus$. We can assume that newly formed LSW contained the same $\text{^{14}C}$ concentration as Labrador Sea surface water because the strong winter convection process homogenizes the surface and intermediate depth waters (Smith et al., 1997). One primary pathway of LSW is a southward outflow through Orphan Knoll (Pickart et al., 2003, Sy et al., 1997). The average transportation rate of outflow in this western boundary current is around 6.6 cm s$^{-1}$ (Lavender et al., 2000), and so it only takes about 1 year for LSW to arrive at Orphan Knoll from Labrador Sea (Pickart et al., 2003). Compared to the 400 year reservoir age of modern high latitude...
North Atlantic Ocean surface waters, the transportation time of LSW from its formation site to Orphan Knoll is negligible. Hence, we assume that in the past cold-water corals at Orphan Knoll remain in a western boundary current and record Labrador Sea surface \(^{14}\text{C}\) information and can be used as an archive to reconstruct paleo-reservoir age of high latitude North Atlantic Ocean. The cold-water coral *D. dianthus* grew in abundance on Orphan Knoll during the last deglaciation (Smith et al., 1997). The abundance of dropstones and steep escarpments at Orphan Knoll apparently provide optimum attachment sites for *D. dianthus* (Schumacher and Zibrowius, 1985). The cold-water coral *D. dianthus* inhabit depths ranging from 60 - 4000 m, but are reported mostly between 500 and 2000 m possibly due to hard substrate availability and the increased solubility of aragonite at deeper depth. *D. dianthus*, construct skeletons of aragonite, and grow at vertical extension rates ranging from 0.5 mm/yr to 2 mm/yr (Adkins et al., 2004, Risk et al., 2002). The high U content of cold-water coral (~ 4 ppm) makes it suitable for \(^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}\) dating, to yield an independent calendar age (Cheng et al., 2000, Goldstein et al., 2001, Schroder-Ritzrau et al., 2003, Smith et al., 1997). Smith et al., (1997) were first to report \(^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}\) dating of *D. dianthus* specimens and demonstrated the suitability of cold-water corals for paleoceanographic reconstructions. Six cold-water coral *D. dianthus* samples were physically and ultrasonically cleaned in Milli-Q water to remove the dark MnO\(_2\) and Fe-oxides/hydroxides coatings. After the physical cleaning process, we applied a chemical cleaning process (Cheng et al., 2000) to remove exterior contaminants. All samples were screened under a binocular microscope for evidence of organic coatings or iron-manganese oxide crusts or various diagenetic
features such as micro-borings, encrustations, etc. before $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ dating and $^{14}\text{C}$ dating.

We determined the $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ ages of cold-water corals by multi-collector magnetic sector double-focusing Inductively Coupled Mass Spectrometry (FIONS PLASMA 54) (Mortlock et al., 2005). $^{14}\text{C}$ dates were measured at Lawrence Livermore National Lab using Accelerator Mass Spectrometry (CAMS) (Table 1, Figure 1).

The six cold-water coral samples yield $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ ages ranging from $\sim 12,800$ to $\sim 13,800$ cal year BP (Table 1, Figure 1). U concentrations range between 3.9 and 5.4 ppm (Table 1), similar to those measured in modern and other fossil cold-water corals (Cheng et al., 2000, Goldstein et al., 2001, Schroder-Ritzrau et al., 2003, Smith et al., 1997). The initial $\delta^{234}\text{U}$ values averaged $149 \pm 1$ per mil (1 SD; $n = 6$), and are in the range of recent measurements of the $\delta^{234}\text{U}$ of present-day seawater (Delanghe et al., 2002). Our cold-water coral initial $\delta^{234}\text{U}$ values are slightly higher than the initial $\delta^{234}\text{U}$ measured for the modern surface corals and within error of reported values for modern cold-water corals of $145.9 \pm 0.8$ (1 SD; $n = 8$), and modern surface water corals $145.3 \pm 2.3$ ‰ (2 SD; $n = 20$) (Cheng et al., 2000, Mortlock et al., 2005), respectively. The consistency of the initial $\delta^{234}\text{U}$ values of our cold-water fossil corals with initial $\delta^{234}\text{U}$ values of seawater and modern corals suggest that diagenesis is negligible.

In contrast to U concentrations, both initial $^{230}\text{Th}$ concentration and $^{232}\text{Th}$ concentration in cold-water coral are much higher than in surface corals (Cheng et al., 2000, Cutler et al., 2004). There are two possible sources of contaminating $^{230}\text{Th}$. First, the iron-manganese oxide crust outside coral adsorbs thorium from seawater (Cheng et al., 2000). We presumably removed the contaminating $^{230}\text{Th}$ by applying a combined physical and
chemical cleaning technique to remove the Mn-Fe oxide coating before the
$^{230}$Th/$^{234}$U/$^{238}$U dating. Second, since deep ocean waters have higher thorium
concentration than surface waters (Guo et al., 1995, Moran et al., 1995), elevated $^{230}$Th
(compared to the surface ocean) was incorporated into the aragonite during the growth of
the coral.

We corrected for the initial $^{230}$Th in our $^{230}$Th/$^{234}$U/$^{238}$U age calculation by calculating
the initial $^{230}$Th/$^{232}$Th activity ratio from $^{234}$U/$^{232}$Th vs. $^{238}$U/$^{232}$Th, and $^{230}$Th/$^{232}$Th vs.
$^{238}$U/$^{232}$Th diagrams (Fig 3). We assume that all our cold-water corals D. dianthus have
the same initial $^{230}$Th/$^{232}$Th activity ratio because they are the same coral species taken
from the same sample location, and three (OK3, OK4, and OK8) of the six samples are
sub-samples from the same coral bouquet, while the others are all close in age. We
plotted the $^{234}$U/$^{232}$Th activity ratio and $^{230}$Th/$^{232}$Th activity ratio vs. $^{238}$U/$^{232}$Th activity
ratio of our samples, respectively (fig 3). The slope of $^{234}$U/$^{232}$Th activity ratio vs.
$^{238}$U/$^{232}$Th activity ratio trend line is the estimated $\delta^{234}$U$_{initial}$ value of our cold-water
corals. The value of the slope is 1.135, which is very close to the seawater $\delta^{234}$U value,
and supports our assumptions described above and indicates that our cold-water corals
formed in a closed system. We then use the y-intercept of the $^{230}$Th/$^{232}$Th activity ratio vs.
$^{238}$U/$^{232}$Th activity ratio regression line as the initial $^{230}$Th/$^{232}$Th activity ratio of our cold-
water corals. The diagrams also allow us to identify potential problematic samples by
checking the data fit to the regression lines. Samples that may have violated assumptions
of equal initial $^{230}$Th/$^{232}$Th should plot off of the trend although none did in this data set.

Using age equations reported in Ivanovich et al. (Ivanovich et al., 1992) and a value of
13.74 ± 8.72 for the $^{230}$Th/$^{232}$Th initial activity ratio, we calculated the corrected
$^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ ages of our cold-water corals (Table 1). The corresponding radiocarbon dates were measured at Lawrence Livermore National Laboratory (LLNL) Center for Accelerator Mass Spectrometry (CAMS) and are listed in Table 2.

2.3 Reservoir age estimation and comparison between reservoir age derived from cold-water corals and other archives

Our cold-water coral data fall in the Allerod (12 890-14 010 cal year BP) and YD (11 650-12 890 cal year BP) climate intervals according to their calendar age assignments reported in Stuiver et al., (1995) (Fig 4, Table 1). The reservoir age estimations ($^{14}$C age differences between the atmospheric and marine data in the same calendar age) are shown in Figure 4 and Table 3. Figure 4 and Table 3 also combines reservoir age estimations using fossils deposited contemporaneously with the age of the Vedde Ash Bed (Austin et al., 1995, Bard et al., 1994, Bondevik et al., 2001), and reservoir age estimations using coexisting terrestrial material and marine shell fragments (Bjorck et al., 1998, Bondevik et al., 1999, Bondevik et al., 2006). The reservoir age estimations from our western North Atlantic and central and eastern North Atlantic Ocean at different locations and various archives generally agree. During the Bolling and Allerod warm period, the averaged reservoir age (380 ± 140 year, n = 14) was indistinguishable from modern values. Although there may be high frequency variability of reservoir age in the Allerod, the reservoir age errors are too large to verify this possibility. At the transition from the Allerod warm period to the YD cold event, the reservoir age was 200 years higher than modern value, and remained close to this value through at least the middle of the YD. The averaged value for the entire YD cold event was 590 ± 130 year, (n = 10).
After the YD cold event at the early Preboreal event, the reservoir age (270 ± 20 year, n = 2) decreased significantly although the data are too sparse to make a reliable estimate.

3. Discussion

Previous reservoir age reconstructions were mostly obtained from coastal sites in the eastern high latitude North Atlantic (Bard et al., 1994, Bjorck et al., 1998, Bondevik et al., 1999, Bondevik et al., 2006, Bondevik et al., 2001), except for one measurement obtained in the central high-latitude North Atlantic (Waelbroeck et al., 2001). Our cold-water coral results span the deglacial Allerod and YD intervals of the western high latitude North Atlantic. For comparison, our reservoir age estimations are consistent with coastal reservoir age reconstructions in the eastern high latitude North Atlantic based on paired marine and terrestrial dated samples. The similar reservoir age estimates between the eastern high latitude North Atlantic and western high latitude North Atlantic provides support for the composite reservoir age plot derived from these two approaches.

Waelbroeck et al., (2001) reported an 800 to 1000 year reservoir age at the end of YD for high latitude North Atlantic by applying the GRIP ice core chronology based on comparison of the δ¹⁸O record (Johnsen et al., 1997) to their deep-sea core δ¹⁸O record. They adopted model results to explain the high and variable reservoir age results, and concluded that reduction in NADW overturning and sea-ice extension caused the reservoir age increase. We chose not to include Waelbroeck et al., (2001) reservoir age estimates in our summary figure 4, because we believe the reservoir age estimation based on imported ice core chronologies contains large sources error, depending upon which ice core age model (Andersen et al., 2004, Grootes and Stuiver, 1997, Johnsen et al., 1997, Shackleton et al., 2004, Southon, 2004, Stuiver et al., 1995), the tie points used, and the
synchronicity of the two proxies at and between the tie points. Furthermore, the reservoir age increases predicted by models of varying complexities (Butzin et al., 2005, Stocker, 1998, Stocker and Wright, 1996) are much lower than reservoir ages reported by Waelbroeck et al., (2001). Coupled ocean-ice-atmosphere model results indicated that the reduction of NADW production and increased ice cover can lead to a 200 to 300 year surface ocean reservoir age increase in the high latitude North Atlantic region (Stocker, 1998, Stocker and Wright, 1996). Another model result showed a 400 to 500 year reservoir age increase by decreasing tropic wind speed and slowing-down the ocean circulation (Delaygue et al., 2003). Fundamentally, the models simulate the response of NADW to meltwater discharge. The model simulated reservoir age result indicate ~100 year fluctuations in the process of meltwater discharge, while the reservoir age increased instantaneously at the end of meltwater discharge due to the resumption of NADW production and deep mixing with $^{14}$C-depleted subsurface waters. However, no meltwater discharge has been identified with the YD (Lowell et al., 2005) despite the modeling activities.

Bondevik et al., (2006) published a North Atlantic radiocarbon reservoir age record during the Bolling / Allerod and YD event based on marine and terrestrial deposits. Bondevik et al., (2006) suggested a high reservoir age up to 500 to 600 years at the end of Allerod period, followed by a rapid decrease to 400 years at the Allerod / YD transition, then the reservoir age increased again gradually to 600 year in the middle YD. Our composite figure includes paired terrestrial/marine $^{14}$C dates from Bondevik et al., (2006), but we do not use the points where only marine $^{14}$C was measured and the terrestrial $^{14}$C was interpolated because of the uncertainties in this
approximation. Our composite results suggest a 400 year reservoir age during the Bolling and Allerod period, and around a 600 year reservoir age through the entire YD cold event. At the end of Allerod period (13000 to 13400 year BP), we had relatively large variations at the reservoir age estimates, from 600 years to 300 years with large error bars from different archives. Due to the relatively small change and correspondingly large uncertainty in the estimates, we are not able to resolve the exact timing of this important transition to increased reservoir ages from the mid-Allerod to the YD.

Bard et al., (1994), Bondevik et al., (1999), and Bondevik et al., (2006) suggested that the reduced NADW production associated with reduced advection of surface inflow water to the North Atlantic corresponds to the increase of reservoir age at high latitude North Atlantic during the YD. In combination with possibly reduced NADW production in the mid-Allerod and YD, the results reported in Rickaby and Elderfield (2005) indicate a possible increased mixture of $^{14}$C depleted AAIW into the high latitude North Atlantic Ocean. Due to its Southern origin, AAIW is expected to have a much lower $^{14}$C concentration than North Atlantic surface water. The modern $^{14}$C “age” of AAIW is roughly 800 to 1000 year (Matsumoto and Key, 2004). Based on the low glacial $\delta^{13}$C values for the Southern Ocean intermediate and deep waters (Charles and Fairbanks, 1992), we assume that AAIW had relatively low $^{14}$C concentrations during the last glacial and deglacial intervals. It has been generally accepted that deep ocean circulation was reorganized and the volume of NADW was reduced during the LGM (Boyle and Keigwin, 1985, Curry and Oppo, 2005, Duplessy et al., 1988, Mix and Fairbanks, 1985, Piotrowski et al., 2005, Sarnthein et al., 2000). In the North Atlantic upper ocean, a shallower Glacial North Atlantic Intermediate Water (GNAIW) replaced NADW (Charles and Fairbanks,
1992, Curry and Oppo, 2005, Duplessy et al., 1988, Oppo and Lehman, 1993). While in the deep ocean, lower $\delta^{13}C$ and higher Cd/Ca in benthic foraminiferal showed that AABW penetrated further north (Boyle and Keigwin, 1985, Curry and Oppo, 1997). An important characteristic of glacial North Atlantic circulation was that glacial North Atlantic was strongly stratified (Duplessy et al., 1988, Oppo and Lehman, 1993). The distribution of $\delta^{13}C$ and Cd data suggest that AABW was present only below 2000 m in the North Atlantic ocean (Bertram et al., 1995), which suggests that it is unlikely that upwelling of old AABW increased the North Atlantic ocean surface reservoir age. Recently, Rickaby et al., (2005) suggested large variations in AAIW in the North Atlantic during the last deglacial based on their benthic foraminiferal $\delta^{13}C$ and Cd/Ca record from deep-sea core NEAP 4K (61.30°N, 24.15°W, 1627m) (Fig 4). The $\delta^{13}C$ was ~1.0 per mil at early Bolling and Allerod warm intervals and early Holocene, while it showed a low $\delta^{13}C$ excursion (up to 0.5 per mil) right before the start of YD and lasted into the YD. This low $\delta^{13}C$ excursion implies that AAIW reached high into the North Atlantic Ocean where deep winter storm mixing can mix AAIW with surface waters. The match of the low $\delta^{13}C$ excursion and the high reservoir age during the YD suggests that AAIW may have contributed to the increase of reservoir age in the end of Allerod and during the YD. Other cold-water coral studies showed the $^{14}C$ enriched and depleted waters fill in the western North Atlantic intermediate depth alternatively to support the invasion of AAIW during the last deglacial (Adkins et al., 1998, Robinson et al., 2005). Siani et al., (2001) previously suggested that southern source water might be responsible for an ~ 800 year reservoir age during the Heinrich 1 event in the Mediterranean Sea. It is possible that the contribution of AAIW was greater at Orphan Knoll than in the high latitude North
Atlantic surface waters. However, the consistency of our cold-water corals estimates of surface water reservoir age and the estimates from entirely independent methods supports our assumption that Orphan Knoll water was rapidly exchanged with high latitude North Atlantic surface water.

Furthermore, we derived atmospheric $\Delta^{14}C$ record from Fairbanks 0805 calibration curve to further infer the intermediate and deep ocean circulation at high latitude North Atlantic Ocean (Fig 4). The record of atmospheric $\Delta^{14}C$ retains a history of solar production changes, carbon cycle perturbations, including changes in the deep-water production, the size of major carbon reservoir, and their exchange rate. Our atmospheric $\Delta^{14}C$ record fluctuated rapidly from 200 to 240 per mil during the Bolling-Allerod warm period, and increased from ~210 per mil to ~250 per mil during the YD transition, when the NGRIP $\delta^{18}O$ record decreased from ~38 per mil to ~42 per mil. The atmospheric $\Delta^{14}C$ increase during YD transition has been reported previously (Edwards et al., 1993, Goslar et al., 2000, Hughen et al., 2000), but the amplitude in our record is smaller and more consistent with model simulations (Delaygue et al., 2003, Stocker, 1998, Stocker and Wright, 1996). Model results suggest that the elevated atmospheric $\Delta^{14}C$ was associated with rapid NADW reduction at the beginning of YD.

4. Conclusion

The reservoir age of North Atlantic high latitude ocean surface water was computed from the radiocarbon age difference between the atmosphere and marine records. We estimate that the reservoir age was similar to the present day value during the Bolling and early Allerod period. From the beginning of YD cold event, the North Atlantic surface ocean $^{14}C$ reservoir age is estimated to have increased to 600 ~ 700 year, and maintained
these high values through the YD cold event. The reservoir age decreased 300 years to values close to modern values at early Preboreal. The high reservoir age at the YD/Allerod transition is consistent with the timing and amplitude of the 40 per mil atmospheric $\Delta^{14}C$ increase. $^{14}C$ reservoir variations may have been caused by variations of AAIW reaching the high latitude North Atlantic Ocean. The inferred role of AAIW in regulating the high latitude North Atlantic surface $^{14}C$ reservoir age and the rapidity of its change builds upon the growing evidence that AAIW plays a critical role in rapid climate change.

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Fig 1 Atmospheric and marine radiocarbon record. Atmospheric record includes surface corals (grey solid circle) (Fairbanks et al., 2005), tree rings (grey cross) (Friedrich et al., 2004) and re-anchored floating tree rings (grey solid triangle) (Kromer et al., 2004). Marine radiocarbon record includes cold-water corals from Orphan Knoll (red open circle), co-existing marine and terrestrial materials (green open square) (Bondevik et al., 1999), (black open triangle) (Bjorck et al., 1998), (blue open diamond) (Bondevik et al., 2006), and Vedde ash layer (grey open star) (Austin et al., 1995, Bard et al., 1994, Bondevik et al., 2001). The $^{14}$C ages of marine samples are raw data, without marine reservoir age correction. We converted the radiocarbon age of terrestrial material and the Vedde ash layer to calendar age using our radiocarbon calibration curve (Fairbanks 0805) (Fairbanks et al., 2005). Each data point is plotted with $1\sigma$ error bars, except tree rings, floating tree rings, and Vedde ash data from Austin et al., (1995) and Bard et al., (1994). The curve segment is computed using a hierarchical Bayesian statistical model, plotted with $1\%$ confidence limit (Fairbanks et al., 2005).
Fig 2. Map of the high latitude North Atlantic Ocean showing current surface and deep ocean circulation. Red dots are locations of cold-water coral sample site Orphan Knoll, other sample locations for reservoir age estimations listed in figure 4 and table 3, and NEAP 4K, which is the benthic foraminifera $\delta^{13}C$ record deep-sea core location. The crosses are approximate locations of deep-water formation. The yellow lines show the southward North Atlantic Drift (NAD), the white dash lines show the pathway of Labrador Sea Water (LSW), and the blue line shows the North Atlantic Deep Water (NADW).
Fig 3. Uranium-series isotope activity ratio diagrams used for estimating initial $^{230}\text{Th}/^{232}\text{Th}$ activity ratio of our cold-water corals. All the ratios showing in diagrams are activity ratios. The y intercept of $^{230}\text{Th}/^{232}\text{Th}$ vs. $^{238}\text{U}/^{232}\text{Th}$ regression line is $13.74 \pm 8.72$, which used as initial $^{230}\text{Th}/^{232}\text{Th}$ activity ratio of our cold-water corals. We applied origin 6.1 software to do the linear regression and error propagation.
Fig 4. From bottom to top are reservoir age estimations, $\delta^{13}C$ record obtained from benthic foraminifera from high latitude North Atlantic deep-sea core NEAP 4K (61.30°N, 24.15°W) (Rickaby and Elderfield, 2005), atmospheric $\Delta^{14}C$ record derived from Fairbanks 0805 calibration curve (Fairbanks et al., 2005), and NGRIP $\delta^{18}O$ record (Rasmussen et al., 2006). Reservoir age estimation obtained by dating cold-water coral, co-existing marine and terrestrial materials (Bjorck et al., 1998, Bondevik et al., 1999, Bondevik et al., 2006), and Vedde ash layer (Austin et al., 1995, Bard et al., 1994, Bondevik et al., 2001). Symbols in this plot are same as symbols in Figure 1. We calibrated the AMS $^{14}C$ dates of the $\delta^{13}C$ record using Fairbanks 0805 calibration curve (Fairbanks et al., 2005). The climate boundaries are defined using ice core chronology ages assigned to characteristic $\delta^{18}O$ variations in GISP2 ice core (Stuiver et al., 1995). The vertical grey shading marks the transition from Allerod warm period to YD cold event.
Table 1. U and Th isotopic composition and $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ ages of cold-water corals.

All errors are 1$\sigma$. We corrected $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ age using an initial $^{230}\text{Th}/^{232}\text{Th}$ activity ratio of 13.74 ± 8.72. Calendar age uncertainty is a combination of the uncertainty in the initial $^{230}\text{Th}/^{232}\text{Th}$, and measured isotope ratio uncertainties. Numbers in brackets are isotope activity ratios.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>U (ppm)</th>
<th>$^{232}\text{Th}$ (ppt)</th>
<th>[230$\text{Th}/^{232}\text{Th}$]</th>
<th>[238$\text{U}/^{232}\text{Th}$]</th>
<th>[234$\text{U}/^{232}\text{Th}$]</th>
<th>$\delta^{234}\text{U}$ (meas.)</th>
<th>$\delta^{234}\text{U}$ (ini.)</th>
<th>Corr. Age (year BP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OK 3</td>
<td>3.9201 ± 0.0021</td>
<td>1171 ± 3</td>
<td>1367 ± 3</td>
<td>10236 ± 24</td>
<td>11621 ± 15</td>
<td>144 ± 0.5</td>
<td>149 ± 1</td>
<td>13375 ± 123</td>
</tr>
<tr>
<td>OK 4$^a$</td>
<td>5.3522 ± 0.0058</td>
<td>4078 ± 12</td>
<td>556 ± 2</td>
<td>4011 ± 13</td>
<td>4555 ± 7</td>
<td>144 ± 0.5</td>
<td>150 ± 1</td>
<td>13687 ± 398</td>
</tr>
<tr>
<td>OK 8</td>
<td>3.9802 ± 0.0015</td>
<td>2550 ± 3</td>
<td>633 ± 1</td>
<td>4771 ± 5</td>
<td>5422 ± 4</td>
<td>145 ± 0.5</td>
<td>150 ± 1</td>
<td>13098 ± 203</td>
</tr>
<tr>
<td>OKB B3</td>
<td>4.2620 ± 0.0013</td>
<td>1618 ± 2</td>
<td>1080 ± 1</td>
<td>8053 ± 7</td>
<td>9139 ± 6</td>
<td>143 ± 0.5</td>
<td>149 ± 0.5</td>
<td>13401 ± 114</td>
</tr>
<tr>
<td>OKB 53B</td>
<td>4.4243 ± 0.0013</td>
<td>5828 ± 6</td>
<td>330 ± 1</td>
<td>2320 ± 3</td>
<td>2632 ± 2</td>
<td>143 ± 0.5</td>
<td>148 ± 1</td>
<td>13847 ± 98</td>
</tr>
<tr>
<td>OKB 36A</td>
<td>4.0233 ± 0.0018</td>
<td>5238 ± 4</td>
<td>314 ± 1</td>
<td>2348 ± 2</td>
<td>2667 ± 2</td>
<td>144 ± 0.5</td>
<td>150 ± 1</td>
<td>12886 ± 132</td>
</tr>
</tbody>
</table>

$^a$The isotopic ratio of sample OK4 is used for the initial $^{230}\text{Th}/^{232}\text{Th}$ activity ratio calculation, but not included in the reservoir age calculation, due to the error on the $^{230}\text{Th}/^{234}\text{U}/^{238}\text{U}$ age.
Table 2. Radiocarbon data for cold-water corals. All errors are 1σ.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Lab ID</th>
<th>Fraction modern</th>
<th>$\Delta^{14}$C (per mil)</th>
<th>Conventional raw $^{14}$C age (year BP)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OK 3</td>
<td>CAMS 108655</td>
<td>0.2296 ± 0.0011</td>
<td>-770.4 ± 1.1</td>
<td>11820 ± 40</td>
</tr>
<tr>
<td>OK 4</td>
<td>CAMS 108656</td>
<td>0.2207 ± 0.0010</td>
<td>-779.3 ± 1.0</td>
<td>12135 ± 40</td>
</tr>
<tr>
<td>OK 8</td>
<td>CAMS 108657</td>
<td>0.2313 ± 0.0012</td>
<td>-768.7 ± 1.2</td>
<td>11760 ± 45</td>
</tr>
<tr>
<td>OKB B3</td>
<td>CAMS 112209</td>
<td>0.2206 ± 0.0008</td>
<td>-779.4 ± 0.8</td>
<td>12140 ± 30</td>
</tr>
<tr>
<td>OKB 53B</td>
<td>CAMS 112208</td>
<td>0.2171 ± 0.0008</td>
<td>-782.9 ± 0.8</td>
<td>12270 ± 30</td>
</tr>
<tr>
<td>OKB 36A</td>
<td>CAMS 112207</td>
<td>0.2343 ± 0.0009</td>
<td>-765.7 ± 0.9</td>
<td>11655 ± 35</td>
</tr>
</tbody>
</table>
Table 3. Radiocarbon reservoir age computed from contemporaneous atmospheric and marine radiocarbon ages. Using Fairbanks et al., (2005) radiocarbon calibration program to compute calendar age of terrestrial and marine material from deep-sea core. All errors are 1σ.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Calendar age (year BP)(^a)</th>
<th>Conventional raw (^{14})C age (year BP)(^b)</th>
<th>Conventional atmospheric (^{14})C age (year BP)(^b)</th>
<th>Radiocarbon reservoir age (year)(^d)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold-water coral <em>D. dianthus</em> (this study), Orphan Knoll (50.43°N, 46.37°W)</td>
<td>OKB 36A</td>
<td>12886 ± 132</td>
<td>11655 ± 35</td>
<td>10961 ± 175</td>
</tr>
<tr>
<td>OK 8</td>
<td>13098 ± 203</td>
<td>11760 ± 45</td>
<td>11167 ± 231</td>
<td>593 ± 276</td>
</tr>
<tr>
<td>OKB B3</td>
<td>13401 ± 114</td>
<td>12140 ± 30</td>
<td>11501 ± 264</td>
<td>639 ± 294</td>
</tr>
<tr>
<td>OK 3</td>
<td>13375 ± 123</td>
<td>11820 ± 40</td>
<td>11470 ± 262</td>
<td>350 ± 302</td>
</tr>
<tr>
<td>OKB 53B</td>
<td>13847 ± 98</td>
<td>12270 ± 30</td>
<td>11963 ± 216</td>
<td>307 ± 246</td>
</tr>
<tr>
<td>Molluscan samples in Vedde Ash Bed (Austin et al., 1995), VE 57-09-46, Hebridean Shelf of northwest Scotland</td>
<td>12089</td>
<td>11000</td>
<td>10300</td>
<td>700</td>
</tr>
<tr>
<td>Shell fragment in Vedde Ash Bed (Bondevik et al., 2001), Kvaltjern, Western Norway</td>
<td>12107 ± 100</td>
<td>10920 ± 24</td>
<td>10310 ± 50</td>
<td>610 ± 74</td>
</tr>
<tr>
<td>Contemporaneous marine and terrestrial material (Bjorck et al., 1998), Lake Madljarn, Southwest Sweden</td>
<td>12808 ± 93</td>
<td>11155 ± 130 (835 – 845)</td>
<td>10880 ± 74(^c) (835 – 845)</td>
<td>275 ± 204</td>
</tr>
<tr>
<td></td>
<td>12703 ± 92</td>
<td>11341 ± 134(^c) (845 – 855)</td>
<td>10750 ± 100 (845 – 855)</td>
<td>591 ± 234</td>
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<tr>
<td></td>
<td>12582 ± 439</td>
<td>11305 ± 140 (855 – 865)</td>
<td>10680 ± 350 (855 – 865)</td>
<td>625 ± 490</td>
</tr>
<tr>
<td></td>
<td>13540 ± 208</td>
<td>11960 ± 195 (875 – 885)</td>
<td>11630 ± 190 (875 – 885)</td>
<td>330 ± 385</td>
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<tr>
<td></td>
<td>13384 ± 159</td>
<td>11668 ± 82(^c) (885 – 895)</td>
<td>11470 ± 135 (885 – 895)</td>
<td>198 ± 217</td>
</tr>
<tr>
<td>Contemporaneous marine and terrestrial material (Bondevik et al., 1999), Kulturmyra, Western Norway</td>
<td>13056 ± 107</td>
<td>11445 ± 55 (593 – 597)</td>
<td>11125 ± 65 (593 – 597)</td>
<td>320 ± 120</td>
</tr>
<tr>
<td></td>
<td>12996 ± 106</td>
<td>11565 ± 45 (608 – 610)</td>
<td>11065 ± 60 (609 – 611)</td>
<td>500 ± 105</td>
</tr>
<tr>
<td></td>
<td>13508 ± 133</td>
<td>11825 ± 55</td>
<td>11600 ± 90</td>
<td>225 ± 145</td>
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<tr>
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<td>-------------</td>
<td>-------------</td>
</tr>
<tr>
<td>13524 ± 120</td>
<td>12015 ± 60</td>
<td>11615 ± 70</td>
<td>400 ± 130</td>
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<tr>
<td></td>
<td>(635 – 637)</td>
<td>(635 – 639)</td>
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<tr>
<td>13805 ± 121</td>
<td>12105 ± 60</td>
<td>11905 ± 85</td>
<td>200 ± 145</td>
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<tr>
<td></td>
<td>(659 – 660)</td>
<td>(659 – 661)</td>
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</tr>
<tr>
<td>13718 ± 119</td>
<td>12335 ± 65</td>
<td>11810 ± 70</td>
<td>525 ± 135</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(672 – 675)</td>
<td>(672 – 675)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>13774 ± 109</td>
<td>12250 ± 50</td>
<td>11870 ± 60</td>
<td>380 ± 110</td>
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<tr>
<td></td>
<td>(683 – 688)</td>
<td>(683 – 688)</td>
<td></td>
<td></td>
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<tr>
<td>14280 ± 202</td>
<td>12685 ± 55</td>
<td>12315 ± 95</td>
<td>370 ± 150</td>
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<tr>
<td></td>
<td>(721 – 722)</td>
<td>(720 – 726)</td>
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</tr>
</tbody>
</table>

Contemporaneous marine and terrestrial material (Bondevik et al., 2006), Kvaltjern, Western Norway

<table>
<thead>
<tr>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>11262 ± 67</td>
<td>10120 ± 60</td>
<td>9865 ± 70</td>
<td>255 ± 130</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(716.5)</td>
<td>(716.5)</td>
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<tr>
<td>11310 ± 96</td>
<td>10195 ± 60</td>
<td>9915 ± 65</td>
<td>280 ± 125</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(740.5)</td>
<td>(740.5)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12022 ± 122</td>
<td>10780 ± 40</td>
<td>10260 ± 65</td>
<td>520 ± 105</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(802)</td>
<td>(802)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12586 ± 63</td>
<td>11125 ± 70</td>
<td>10590 ± 65</td>
<td>535 ± 135</td>
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</tr>
<tr>
<td></td>
<td>(871)</td>
<td>(871)</td>
<td></td>
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</tr>
<tr>
<td>12884 ± 107</td>
<td>11615 ± 95</td>
<td>10960 ± 75</td>
<td>655 ± 170</td>
<td></td>
</tr>
<tr>
<td></td>
<td>(908)</td>
<td>(908)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)Calendar age for samples other than cold-water corals are computed by converting the terrestrial radiocarbon age into calendar age using Calibration curve (Fairbanks 0805) (Fairbanks et al., 2005)

\(^b\)The numbers in the parentheses are sample depth in the sediment core. The unit is cm.

We only use paired marine and terrestrial data that follows the stratigraphic sequence for the reservoir age calculation.

\(^c\)These ages are weighted average value of different fraction of terrestrial deposit or marine sediment.

\(^d\)We calculated the errors of reservoir age by summing the error of marine and terrestrial radiocarbon age.